FEDERAL/PROVINCIAL
RESEARCH AND MONITORING
COORDINATING COMMITTEE (RMCC)

ASSESSMENT OF THE STATE OF KNOWLEDGE ON THE LONG-RANGE TRANSPORT OF AIR POLLUTANTS AND ACID DEPOSITION

PART 1
EXECUTIVE SUMMARY

AUGUST 1986

ACID RAIN

PLUIES ACIDES

Province of British Columbia
Ministry of Environment

Gouvernement du Québec
Ministère de l'Environnement

Alberta
ENVIRONMENT

New Brunswick
Department of Municipal Affairs and Environment

Saskatchewan Environment

Prince Edward Island
Community & Cultural Affairs

Manitoba Environment and Workplace Safety and Health

Nova Scotia
Department of the Environment

Ontario
Ministry of the Environment

Province of Newfoundland
Department of the Environment

Canada

Assessment of the state of knowledge on the long-range transport of air pollutants and acid deposition.

FEDERAL/PROVINCIAL RESEARCH AND
MONITORING COORDINATING COMMITTEE (RMCC)

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1.1 INTRODUCTION

In the fall of 1984, the Federal/Provincial Acid Rain Research & Monitoring Coordinating Committee (RMCC) decided to undertake an assessment of the state of knowledge on the long-range transport of airborne pollutants and acid rain. There were several reasons for making the decision at that time, including:

a. there had been no major Canadian assessment of new scientific information since the U.S. - Canada Memorandum of Intent (MOI) activity approximately five years ago,

b. the large commitment of resources by Canadian governments to acid rain studies had yielded a significant amount of new information,

c. the U.S. National Acidic Precipitation Assessment Program (NAPAP) was completing its first assessment of acidification knowledge in the United States, and

d. significant changes had occurred in the acid rain issue including a dramatic demonstration of forest deterioration in Europe, reports of forest decline in North America, a significant increase in our understanding of the responses of aquatic systems to acidification, and the implementation of a control program in Canada.

This assessment does not cover all research results produced since the Memorandum of Intent documents were completed. Instead, the assessment is limited to scientific information which relates to a number of specific questions. In many instances, the questions have direct implications on the policies that the Governments of Canada are implementing in order to deal with this environmental problem. No policy analysis is given here.

The assessment report includes new scientific information from the Muskoka '85 International Symposium on Acid Precipitation where over 400 scientific papers were presented. Many of these papers will be published in the Journal of Water, Air and Soil Pollution in the fall of 1986. The scientific literature provides the remainder of the review material. The MOI documents are referenced from time to time for the purpose of maintaining continuity. Results from research programs outside of Canada are included where appropriate.

The report is made up of six sections, this Executive Summary and five separate parts, viz:

Part 1:	Executive Summary	(17 pages)
Part 2:	Atmospheric Sciences	(108 pages)
Part 3:	Aquatic Effects	(57 pages)
Part 4:	Terrestrial Effects	(80 pages)
Part 5:	Human Health Effects	(26 pages)
Part 6:	Effects on Man-Made Structures	(30 pages)

The Executive Summary draws on the key findings from the other sections. Except for Part 6 on Man-Made Structures, the reports have been written by government scientists. Part 6 was prepared under contract, by Dr. P. Sereda, former head of the Division on Materials Corrosion, National Research Council of Canada.

The names and affiliations of all authors are given in Appendix 1.

Copies of all reports may be obtained from:

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Ces rapports sont aussi disponibles en français.

1.2 ATMOSPHERIC SCIENCES

1.2.1 What are the present emissions of acidifying pollutants in North America? How have emissions changed during the past 30 years?

The most comprehensive emission inventory for SO_2 and NO_X is the base year 1980. Canadian SO_2 emissions increased from 4.6 million tonnes (1955) to 6.7 million tonnes (1970) and dropped to 4.6 million tonnes (1980). Emissions of SO_2 in the U.S. rose from 20 million tonnes (1950) to 28 million tonnes (1970) and dropped to 24 million tonnes (1980) (see Figure 1). 80% of the total emissions come from the 31 states east of the Mississippi River and the provinces east of the Manitoba-Saskatchewan border. From 1979 to 1982 the decline in SO_2 emissions accelerated in both countries, coincident with the economic recession. There is an indication that emissions in North America increased beyond 1983, however, validated Canadian emission data is not yet available.

Canadian NO_X emissions have also more than doubled since the early 1950's, from 0.6 million tonnes in 1955 to 1.7 million tonnes in 1980. Emissions of NO_X in the U.S. have increased significantly from about 9 million tonnes in 1950 to 21 million tonnes in 1980 (see Figure 2). The eastern parts of both countries are responsible for more than 60% of the emissions.

There have been some important changes in the characteristics of emission sources for the past 30 years. In the Northeastern portion of North America there has been a redistribution of SO_2 emissions from small urban sources to large rural sources (power plants) and, in the U.S., an increase in emissions in the south has occurred, associated with a major shift in population to that area. A progressive increase in the proportion of SO_2 released from tall stacks has occurred, enhancing the potential for long-range transport of this gas.

1.2.2 What are the present regional scale concentration/deposition patterns?

In southern and central Ontario, southern Quebec, and over most of the U.S. region east of the Mississippi Valley wet deposition loadings of sulphate exceed 20 kg/ha/yr (see Figure 3). The boundary of this area varies by a few hundred kilometers from year to year with the Atlantic Provinces falling within or outside of the 20 kilogram contour line.

On the west coast, the deposition patterns are not well defined because of the very complex terrain and highly variable precipitation, however, the sulphate deposition in precipitation appears to be below 20 kg/ha/yr. The pH of precipitation is generally well above 5.0 except for some of the regions around the main population centres (i.e. the Los Angeles basin, the San Francisco basin and the Seattle - Vancouver area).

In the Prairie Provinces, wind blown dust plays an important role in determining the chemistry of deposited pollutants. There is no correlation between precipitation pH and SO_4 concentrations because of the confoundings effect of SO_4 derived from the soil components found in the rain. The average rainfall pH is well above 5.6 in the south, but through the north extending into the Yukon and the Northwest Territories, pH values vary between 5.0 and 6.0.

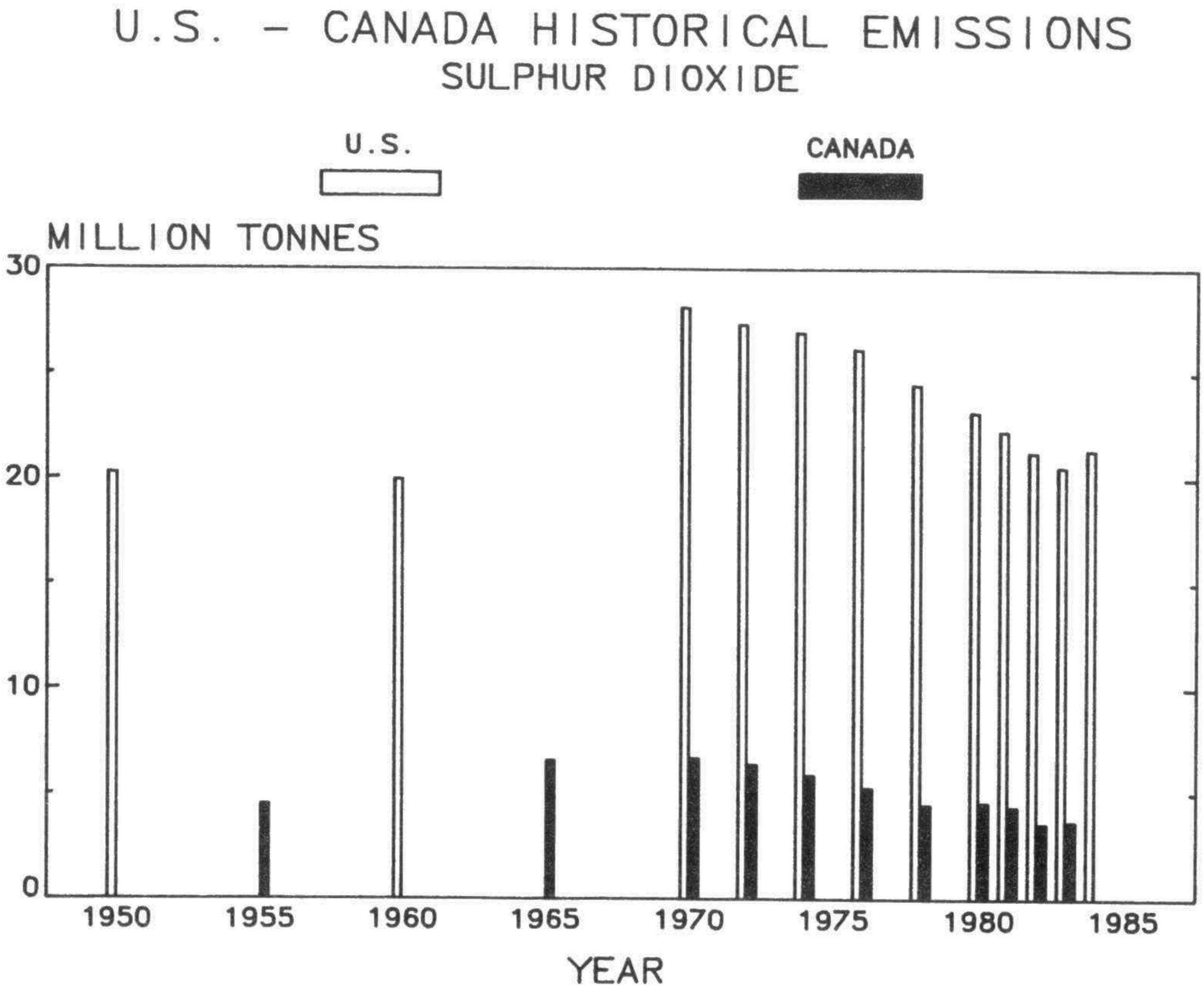
U.S. - CANADA HISTORICAL EMISSIONS
SULPHUR DIOXIDE
U.S.
CANADA
MILLION TONNES
30
20
10
0
1950
1955
1960
1965
1970
1975
1980
1985
YEAR

FIGURE 1

U.S. - CANADA HISTORICAL EMISSIONS
NITROGEN OXIDES

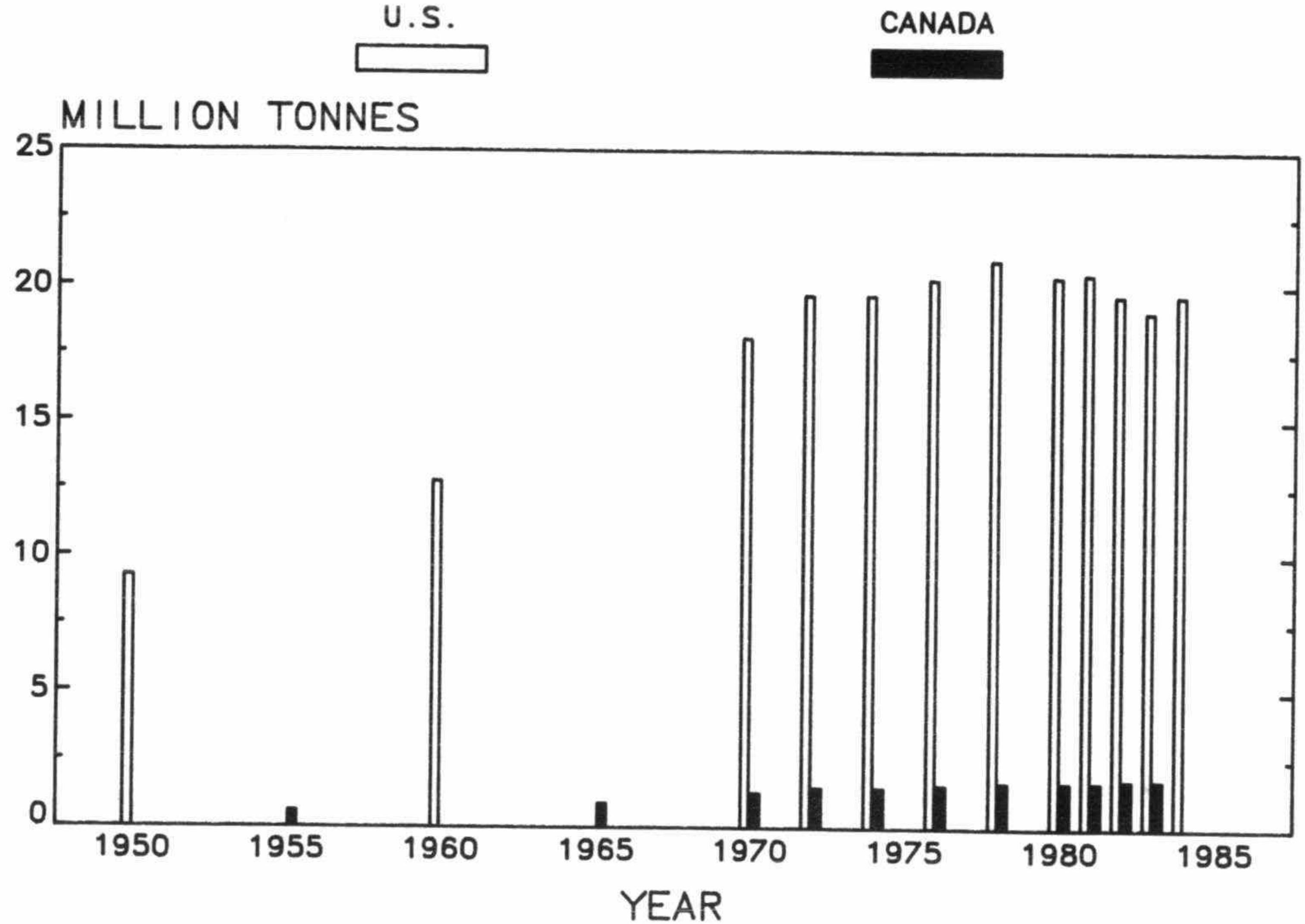

FIGURE 2

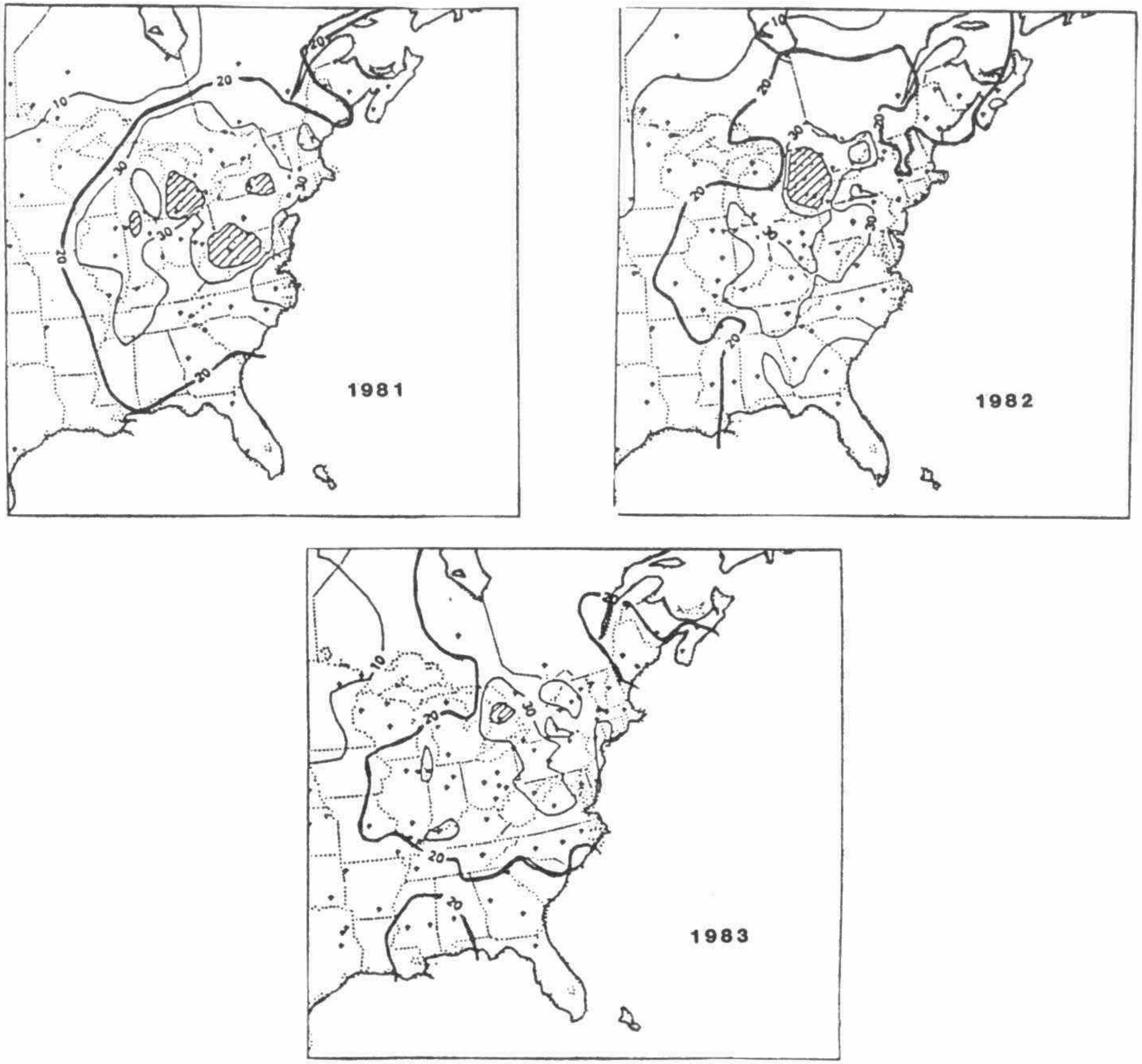

FIGURE 3

ANNUAL SULPHATE DEPOSITION IN PRECIPITATION (kg/ha/yr) IN EASTERN NORTH AMERICA FOR THE YEARS 1981, 1982, 1983.

It is now clear that the composition of wet deposition in the region of heaviest deposition (southeastern Canada and northeast U.S.A.) has a strong annual cycle. There is a maximum of acidity and sulphate in late summer and autumn, a maximum of neutralizing substances such as calcium in the spring, and a maximum of ammonium in autumn. Nitrate deposition tends to be more evenly distributed throughout the year and, hence, contribute relatively more to the acidity of winter snow than to summer rain.

New data have demonstrated that wet deposition of sulphate and nitrate is highly episodic with more than 50% of the annual deposition at eastern stations occurring in only 20% of the precipitation events.

Problems with measuring dry deposition persist. It has been determined, however, that over eastern North America as a whole, dry and wet deposition of sulphur and nitrogen oxides are approximately equal although the relative importance of dry and wet components changes with the distance from major sources. Close to major source regions dry deposition tends to exceed wet.

There is increasing evidence that wet deposition from the direct contact of fog and low clouds with forests at high elevations can provide a deposition of some pollutants which is equivalent in magnitude to either precipitation or dry deposition alone. Cloud water pH values ranging between 3.0 and 4.5 have been recorded with occasional values below 3.0. These new findings have important implications for high elevation forests in some areas.

The acidity of precipitation in Europe and eastern North America is comparable although the concentrations of sulphates in air are higher in Europe. Nitrates play an important role in precipitation chemistry of both continents, with the frequency of occurrence of elevated ozone (a secondary product of NO_x) in Europe being comparable to some areas in the eastern Canada and U.S.A.

1.2.3 Have these patterns changed during the past 30 years?

Changes in the deposition patterns in North America cannot be determined because of the limited availability of records over this period. However, data for the period 1979-83 suggest a recent decrease in sulphate in precipitation in Eastern North America, coincident with decreases in emissions over the same period. In the Eastern U.S.A., particularly the southeast, precipitation is currently more acidic than it was in the mid 1950's and 1960's.

In Europe high quality precipitation data suggest a reasonable correspondence between emissions and deposition over the past 35 years. Deposition in Scandinavia generally follows the estimated emission changes in several of the countries known to contribute to deposition in Scandinavia.

1.2.4 How well can we estimate concentration/deposition fields from source-receptor relationships?

Globally there is a strong geographical association between the regions of highest emissions and of maximum precipitation acidity. The annual wet sulphate deposition in northeastern North America can be up to 16 times greater than in remote regions where deposition levels of 1-3 kgs/hectare/yr are common.

It has been confirmed that, based on simple source-receptor relationships, approximately 50% of the sulphur deposited in eastern Canada has its origin in U.S. sources with the remaining sulphur being derived from Canadian emission sources. Although it is less certain, it appears that nitrogen compounds behave in a similar manner.

At individual stations, the daily measurements of air and precipitation concentrations of nitrates and sulphates are highly variable. Grouping of data by wind direction segments yields a high correlation with emissions within the segments, and observed concentrations. This analytical procedure does not distinguish between nearby and distant sources.

Over the past few years, simple linear transport models have been refined to the point that wet deposition of sulphur over a one-year period is reasonably well simulated, indicating that the non-linear character of sulphur chemistry in the atmosphere is not critical in this application. These models can now be used with considerable confidence in establishing source-receptor relationships for sulphur.

Furthermore, large-scale tracer experiments in eastern North America have confirmed the improved performance of these linear trajectory models under selected meteorological situations and thus have provided additional confidence in the use of the models in estimating source-receptor relationships.

Several Eulerian long-range transport models have now been developed which include a state-of-the-art treatment of atmospheric transport, transformation and deposition processes, thus in a large part addressing the earlier criticisms levelled at the simple numerical models during MOI. These new models have not yet yielded predictions of source-receptor relationships and are still to be validated with monitoring data.

1.3 AQUATIC EFFECTS

1.3.1 What is the evidence that the chemical characteristics of aquatic receptors have changed over the past several decades?

The MOI conclusion that there has been an increase in sulphur and acidity in surface waters of eastern Canada over the past several decades, has been confirmed by recent indirect evidence from sediment core analyses for sulphur, trace metals and diatom fossils. Direct measurement of changes in surface water acidity, alkalinity and sulphate concentrations based on historical records were cited in the MOI documents.

Recent studies show that there has been a decrease in sulphur flux through some aquatic systems since the mid 1970's. In two regions, for which significant records are available (Nova Scotia and the Sudbury area of Ontario), decreases in sulphur fluxes have been associated with increases in alkalinity and pH of surface waters. The decrease in sulphur flux in Nova Scotia is associated with a general reduction in total SO_2 emissions in North America, while the decrease at the Sudbury area is principally a result of a local control program. Findings from both sites support the decision to undertake a sulphur dioxide emission control program in eastern Canada.

For the first time in Canada, there is evidence of short term acidification of shallow groundwater. Studies in the Algoma Region of Ontario and in the Montmorency Forest of Quebec have shown loss of alkalinity of soil water, to a depth of about one metre, associated with elevated concentrations of sulphate and nitrate.

Near Pinawa, Manitoba, a recently completed aquifer study showed a 20-fold increase in sulphate levels in groundwater less than 30 years old compared with water 60 to 240 years old. No pH depression was been observed as these waters are well buffered.

1.3.2 What is the evidence that changes in aquatic chemistry are caused by the deposition of atmospherically transported material?

Across eastern Canada, the annual deposition of sulphate ion is highly correlated with the surface water alkalinity deficit. The correlation between nitrate deposition (or ammonium) and alkalinity is less clear, although during episodic events nitrate increases in importance in some cases.

Organic acids, from within some sensitive aquatic systems, contribute to the alkalinity deficit of some surface waters of eastern Canada but during periods of highest acidity they play a minor role.

Process models which relate aquatic acidification to atmospheric deposition on a watershed scale have been validated for seasonal and monthly periods. Episodic events, particularly those related to snowmelt, are accurately simulated. Longer term modelling (a year or more) has not achieved as great a level of confidence due to the lack of validation of certain critical coefficients, in particular, weathering rates and their spatial variation.

1.3.3 What is the evidence for changes in the aquatic biological community?

Fisheries records of population densities or of fish harvest do not exist for most of the sensitive waters of Canada over sufficient periods to demonstrate direct evidence of changes. However, in southwestern Nova Scotia, loss of the Atlantic Salmon populations have been documented in several rivers and serious decline in harvest have been noted in others. Fish population losses have been documented in the Sudbury area of Ontario and losses and declines have been confirmed in lakes more distant from Sudbury or other strong sulphur emission sources.

Repeated and extensive fish surveys in eastern Canada continue to confirm that species diversity and richness is reduced in acidified surface waters.

Recent evidence shows that recovery of fish populations in the Sudbury area has been coincidental with reduced atmospheric sulphate deposition and improved lake water quality. These findings also support the decision of the Governments of Canada to undertake a sulphur emission control program in the eastern part of the country.

1.3.4 What evidence is there of a relationship between chemical changes and the biological status of the aquatic ecosystem?

In the case of fish, the link between chemical characteristics and biological health has been demonstrated through both increasing and decreasing cycles of acidity. In controlled lake acidification experiments it has been shown that food web disruptions can have significant and, in some cases, devastating effects on the higher order predator species. Reproductive failure also occurs resulting in eventual catastrophic changes in fish populations.

Algae and zooplankton populations decline at pH value below 6. In addition, it has been found that mayflies and common snails are very sensitive to low pH as well as benthic crustaceans which have a critical threshold between pH 5.5 and 6.

It has been conclusively demonstrated that severe depletions of alkalinity (increases in acidity) with associated increases in free aluminum can create water quality conditions toxic to many aquatic organisms. Of particular importance are snowmelt events occurring in the spring when some organisms are in the most vulnerable stage of development. Although, adult fish may avoid lethal conditions if zones of non-toxic waters are available, actual adult kills have been documented and juveniles and lower order species may be severely affected or eliminated.

Field and laboratory experiments have shown that acidic water conditions, particularly waters with pH less than 5.0, are toxic to amphibians.

Recent findings indicate that water fowl breeding success and growth of young may be lower in acidifying streams where prey organisms are limited by nutrient availability as well as pH.

1.3.5 What is the rate of response and rate of return to equilibrium resulting from changing deposition stress levels?

Measurements of surface water chemistry have provided evidence of direct and rapid response to changes in deposition loadings on a short term basis. Long term monitoring for periods in excess of five years are providing regional information. Where the availability of bedrock derived base cations is low (i.e. Canadian Shield), surface water responses to increasing acidic loadings are rapid (within a few months). In regions with higher base cation availability, the decline of surface water alkalinity is slower with delays in the order of many months. There is limited information on the rate of response of both types of systems to decreasing acidic loadings, however studies to date indicate that a fairly rapid reversal in acidification can be expected (years rather than decades).

Rate of response of the biological community remains poorly defined. Recovery may be rapid if extinction has not been reached but will require restocking from other existing gene pools where extinction has occurred.

1.3.6 What are the potential consequences of mitigative measures?

Recent lake pH increases from 5 to over 6 have been achieved at a few lakes in Ontario by the addition of calcium carbonate and calcium hydroxide. Associated decreases in total aluminum concentration have allowed the re-establishment of lake trout populations. On a smaller scale, the application of lime or limestone gravel to spawning shoals has protected trout juveniles.

The liming of rivers in Nova Scotia to protect salmon populations against average acidic conditions and in particular episodic events, has been less successful.

The lake and river liming experiences in Ontario and Nova Scotia have highlighted the problems of full-scale liming operations as a means of rehabilitating and protecting aquatic ecosystems:

1) in highly metal-contaminated systems, liming may not reduce metal concentrations to non-toxic levels,

2) without concommitant reductions in deposition, liming is a continuous process,

3) liming may not protect the habitat for critical developmental periods of all fish species, i.e. during episodic events,

4) the long-term response of resident fish populations to liming has not yet been assesed,

5) on strictly economic terms, full scale liming of Nova Scotia Atlantic salmon rivers cannot be justified,

6) as liming is not an exact reversal of the acidification process and has been studied in a limited number of systems, the response of the resident community cannot be predicted. In Europe liming has occasionally led to domination by non-target or non-desirable species of fish.

1.3.7 What are the aquatic resources at risk?

Estimates from limited surface water surveys indicate that approximately 14,000 Canadian lakes are currently acidified. The model predicts that if present levels of wet deposition in the heavily impacted (greater than 20 kg/ha/yr of wet sulphate) region east of the Manitoba/Ontario border are maintained, a further 10,000 to 40,000 additional lakes will be acidified.

1.3.8 Is the present target loading of 20 kg/ha/yr of wet sulphate protective of the freshwater aquatic ecosystem in eastern Canada?

During the preparation of the Memorandum of Intent Work Group Reports, the target loading of 20 kg/ha/yr of sulphate in precipitation was proposed. The Canadian Acid Rain Control Program for eastern Canada has been based on this recommended sulphate loading, acknowledging that this target will not protect the very sensitive lakes and rivers, a small proportion of the aquatic resource.

The target loading of 20 kg/ha/yr of sulphate in precipitation may not provide protection against episodic acidification events. A sufficient reserve of alkalinity must be maintained in the aquatic system to prevent such events.

Simulation model predictions indicate that a reduction of total sulphate deposition (wet plus dry) to 12 kg/ha/yr may be required to prevent damage to highly sensitive aquatic ecosystems and to protect moderately sensitive lakes and rivers from episodic acidification.

At this time, deposition levels in western Canada are far lower than in the east. The appropriate target loading values for western regions are under review.

1.4 TERRESTRIAL EFFECTS

1.4.1 What is the evidence for LRTAP effects on the terrestrial ecosystem?

In recent years, there has been a marked decline in sugar maple in Quebec and Ontario in areas where there are high loadings of atmospheric pollutants. Along with a wide range of environmental stresses, acid rain has been implicated as a contributing factor to this decline in certain locations.

Visible symptoms and reduction in growth and yield have been associated with ambient ozone concentrations in U.S. forests.

A wide range of laboratory experiments has demonstrated irrefutably that air pollutants including acidic precipitation can influence biochemical and physiological processes in plants. Ambient levels of acid rain and ozone, typical of parts of eastern Canada, have affected forest pollen viability in experimental conditions.

Acidic deposition is known to enhance the leaching of base cations and the solubilization of aluminum in soil, which have negative effects on biological processes. The extent of these changes are being determined on a regional scale.

In some forest ecosystems acidic deposition has increased the cycling of nutrients, in particular, the leaching of foliar base cations.

The impact of ozone alone on crop yield is well documented. In the case of acidic precipitation, however, visual symptoms have not been observed under ambient conditions. Repeated rainfalls with pH less than 3 would be required for crop damage of economic significance.

The evidence to date suggests that air pollution, including acidic precipitation is most likely involved on a long-term basis in the decline of forests in several parts of the world. Direct association is not readily demonstrated since forest ecosystems are subject to multi-stresses including air pollutants, insects, disease, adverse weather, and climate.

1.5 HUMAN HEALTH EFFECTS

1.5.1 What are the direct health effects of long range transport?

The adverse direct effects of high levels of acidic air pollutants have been well documented in both field and laboratory studies. However, it is still unclear whether lower, more normally occurring levels of pollutants are causing either direct or indirect effects.

A 1983 study, using eight years of data, showed an association between increased hospital admissions for respiratory illnesses in southwestern Ontario and increased ambient levels of sulphate, ozone, and temperature. In addition, the incidence of moderate respiratory morbidity (evidenced by either restricted activity and/or outpatient visits to doctor or hospital) was probably higher but was not documented since these data reported only respiratory illness severe enough to warrant hospitalization.

A recent study in central Ontario demonstrated that the levels of fine particles (less than 2.5 microns), average sulphates and maximum daily ozone were significantly associated with a decrement in the lung function of children. The effect was observed in both asthmatics and non-asthmatics.

Another study compared the chronic health effects of exposure to air pollution in school children residing in Tillsonburg, Ontario (a high LRTAP community) and Portage La Prairie, Manitoba (a low pollution community). After adjusting for the confounding influences of parental smoking habits and socioeconomic status, children in Tillsonburg exhibited a small (2%) but statistically significant decrement in lung function as compared with those residing in Portage La Prairie. Children in Tillsonburg also had a higher incidence of the following respiratory symptoms, when compared with their counterparts in Portage La Praire: persistent chest colds (15.4% vs 12%); inhalant allergies (12.6% vs 5.1%); cough with phlegm (25.3% vs 21.6%) and stuffy nose (10% vs 8.2%). The number of children reporting asthma did not differ significantly between the two communities. While levels of NO_x and total respirable particles differed little between the communities, Tillsonburg had overall elevated levels of SO_2, nitrates and respirable sulphates. These findings are suggestive but not conclusive as it is unclear whether observed health differences were due to transported air pollutants or other characteristics which differed between the two communities but were not measured. Further studies to expand and verify these results are currently underway.

1.5.2 What are the indirect health effects of long range transport?

Acid deposition can pose a human health hazard through increases in the levels of toxic components in drinking water by leaching metals from waterbeds and sediments, and by corroding materials used in water distribution and storage systems. Mercury, aluminum, copper, lead, cadmium, and asbestos are contaminants of major concern.

A study conducted in the Muskoka-Haliburton region of Ontario indicated that untreated water standing in pipes over several days can have concentrations of lead, copper, zinc and cadmium which exceed federal guidelines. The concentrations decreased after flushing but remained above source levels, demonstrating that metal concentrations were related to residence time in the pipes.

Currently, studies are being conducted to identify the human population in Canada that obtains its drinking water from non-municipal, unregulated sources.

1.6 EFFECTS ON MAN-MADE STRUCTURES

1.6.1 What part of observed corrosion and deterioration can be attributed to the effect of "acid deposition"?

At this stage the part of observed corrosion and deterioration of the built environment attributed to the effects of acid deposition cannot be estimated. The value of construction in Canada in 1985 was $61 billion (14% of the GNP). Repairs and maintenance were responsible for $11 billion of the total construction costs. Air pollution is one of the agents leading to the required repairs and maintenance.

Our overall ability to define the principal causes of material deterioration and the processes involved is extremely limited. Over the past few years, new information has changed our approach to assessing this consequence of "acid rain". Important new findings include:

- the length of time a material is wet (hours per year) is a critical parameter in determining the amount of corrosion due to air pollution;

- the corrosivity of a given environment is not the same for all metals, hence, the corrosivity of an environment cannot be defined in terms of a single metal but must be defined in terms of the microclimate (physical and chemical properties of air);

- although paints and coatings are perhaps the most important and common component being subjected to deterioration by air pollution, relatively little is known about the process;

- stones comprise a small part of the built environment but a major part of the heritage component. Air pollution, in particular sulphur dioxide gas, plays a dominant role in the deterioration of these materials;

- a building in the urban environment can show a very marked change in corrosion rate with elevation, with the top of the building corroding more rapidly than the lower portion. For this reason, research on individual selected buildings can yield significant information on the overall deterioration of the urban environment;

- the role of precipitation in the corrosion of materials is complicated since rain can be beneficial because of its washing action.

APPENDIX I: AUTHORSHIP OF ASSESSMENT REPORTS

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Part 6: Effects on Man-Made Structures

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